

# Lieb-Mattis ferrimagnetism in diluted magnetic semiconductors

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We show the possibility of long-range ferrimagnetic ordering with a saturation magnetisation of  $\sim 1\mu_B$  per spin for arbitrarily low concentration of magnetic impurities in semiconductors, provided that the impurities form a superstructure satisfying the conditions of the Lieb-Mattis theorem. Explicit examples of such superstructures are given for the wurtzite lattice, and the temperature of ferrimagnetic transition is estimated from a high-temperature expansion. Exact diagonalization studies show that small fragments of the structure exhibit enhanced magnetic response and isotropic superparamagnetism at low temperatures. A quantum transition in a high magnetic field is considered and similar superstructures in cubic semiconductors are discussed as well.

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In order to launch the engineering of a new generation of electronic devices, one needs new materials with special properties. For instance, spintronics has a need for room-temperature ferromagnetic semiconductors<sup>1</sup>. Since the discovery of high- $T_C$  ferromagnetism in GaAs:Mn<sup>2</sup> and the prediction of room-temperature ferromagnetism in  $p$ -doped ZnO:Co,Mn systems<sup>3</sup>, a lot of attempts have been made to obtain ferromagnetism in transition metal doped ZnO, GaN and in other oxides and nitrides. The  $p$ -type carriers doping is necessary for the  $p$ - $d$  Zener ferromagnetic long-range interaction<sup>4</sup>. Up to now all attempts to obtain ZnO with  $p$ -type current carriers have failed. Nevertheless, several reports of “ferromagnetic” room temperature behavior have been published<sup>5-7</sup>. “Perhaps the most surprising development of the past decade in the science of magnetic materials is the abundant observations of spontaneous magnetization persisting to above room temperature in semiconductors and oxides, in which no ferromagnetism was expected at any temperature, particularly in the  $p$ - $d$  Zener model”<sup>5</sup>.

In the absence of  $p$ -type current carriers, the interaction between magnetic impurities is governed by the superexchange mechanism. Superexchange is often regarded as an obstacle in the way towards magnetic semiconductors as it has antiferromagnetic (AFM) character and tends to anti-align the interacting spins, leading to a cancellation of the net magnetization. In fact, the AFM interaction does *not* preclude spontaneous magnetization. In a seminal paper<sup>8</sup>, E. Lieb and D. Mattis showed that the ground state of an AFM system depends on the topology of the interacting bonds and, under certain conditions, it is *ferrimagnetic* rather than AFM. The Lieb-Mattis theorem applies if there is no magnetic frustration in the spin system.

In this communication we study various structures formed by the interacting magnetic impurities in wurtzite semiconductors. We take antiferromagnetic nearest

neighbor interaction into account and consider diluted lattices without frustration, in order to remain within the Lieb-Mattis scheme. First we construct several finite clusters that show an enhanced magnetic response at low temperatures. Not alone do they possess a net magnetic moment, they all share a further interesting peculiarity: below a certain temperature their magnetic susceptibility exceeds that of non-interacting spins. We call it isotropic superparamagnetic response<sup>9,10</sup>. Next we construct extended lattices of these clusters, which undergo a ferrimagnetic ordering transition at a finite temperature. The average ground-state spin per magnetic ion of spin  $S$  tends to a finite value (of about  $S/3$ ) despite the low concentration of magnetic ions. The extension of our idea to other lattices and the influence of frustration will be briefly discussed at the end of the communication.

We take the interaction in the form

$$\hat{H} = \frac{1}{2} \sum_{\mathbf{R}, \mathbf{r}} J_{\mathbf{r}} \hat{\mathbf{S}}_{\mathbf{R}} \hat{\mathbf{S}}_{\mathbf{R}+\mathbf{r}}, \quad (1)$$

i.e., we adopt the notation  $J_{\mathbf{r}}$  for the interaction between one pair of spins<sup>11</sup>. We assume that only the nearest-neighbor (in the metal sublattice) interaction is nonzero. This assumption is relevant to magnetic semiconductors, where the nearest-neighbor exchange dominates<sup>12-14</sup>. Two kinds of nearest neighborships are present in wurtzites: those where both ions lie in the same plane and those where they lie in two adjacent planes. The corresponding exchange integrals,  $J_1$  (in-plane) and  $J_2$  (out-of-plane), are different<sup>15-17</sup>.

The magnetic response of a system is characterized by its magnetic susceptibility. Talking about a compound  $A_{1-x}M_xX$  (where  $X$  is a ligand of V or VI group,  $A$  is a metal of IIId or IIb group, and  $M$  is a transition metal), we shall attribute all the magnetic moment to transition metal ions (TMIs) only. We now introduce the magnetic

susceptibility per one spin,

$$\chi \equiv \frac{\mu_M}{H}, \quad (2)$$

where  $\mu_M$  is the average magnetic moment of one TMI. For non-interacting spins, the susceptibility obeys the Curie law  $\chi_C = [(g\mu_B)^2 S(S+1)]/(3k_B T)$ , where  $S$  is the spin of the TMI and  $g$  is its gyromagnetic ratio. Besides isolated spins, TMI impurities may form pairs, trimers, tetramers, and more complex structures (see Fig. 1). The

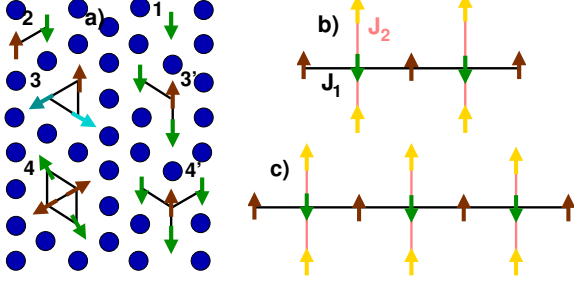


FIG. 1. (Color online) **a)** : Complexes formed by transition metal impurities (arrows): isolated ions (1), dimers (2), trimers (3,3'), tetramers (4,4'). Black solid line segments depict the nearest-neighbor interaction  $J_1$  bonds. One wurtzite  $ab$  plane is shown, blue circles denote non-magnetic host metal ions, ligands are not shown. **b), c)** : More complex Lieb-Mattis systems with ferrimagnetic ground state: linear chains of impurities in the  $ab$  plane "decorated" by spins in adjacent planes (gold arrows); pink line segments depict  $J_2$  bonds.

antiferromagnetic interaction depresses the magnetic response at high temperatures. For  $T \gg J_{max}S(S+1) \equiv T_s$ , the susceptibility of an interacting system obeys the Curie-Weiss law  $\chi_{CW} = [(g\mu_B)^2 S(S+1)]/[3k_B(T-\theta)] < \chi_C$ , with  $-\theta = [S(S+1)]/(3k_B N) \sum_{\mathbf{R}, \mathbf{r}(\mathbf{R})} J_{\mathbf{r}(\mathbf{R})}$ . Here  $N$  is the number of spins and  $J_{max}$  is the strongest exchange interaction in the system,  $\mathbf{R}$  runs over all spins of the lattice, and  $\mathbf{r}$  runs over all nearest neighbors of each spin.

At temperatures  $T \lesssim T_s$ , the response of the system depends on its geometry. Analytic expressions for the susceptibility can be obtained for small systems<sup>18</sup>. Fig. 2a shows the results for the simplest  $S=1/2$  case. We see that at  $T \sim T_s$  the response of three spins arranged linearly **3'** is larger than that of a triangular arrangement of the same spins **3**. For 4-spin systems we see the striking difference between the response of a star arrangement **4'** and that of a rhombus **4**.

Even more interesting is the response of the complexes shown in Fig. 1b,c. Each one of these systems can be decomposed into two sublattices A and B (denoted by arrows "up" and "down"), the interaction being nonzero only between sites that belong to different sublattices. Such a system satisfies the requirements of the Lieb-Mattis theorem<sup>8</sup>, and possesses a ferrimagnetic ground state with total spin  $S_g = S|N_A - N_B|$ . In this case, the term "ferrimagnetic" refers to correlations of the spins

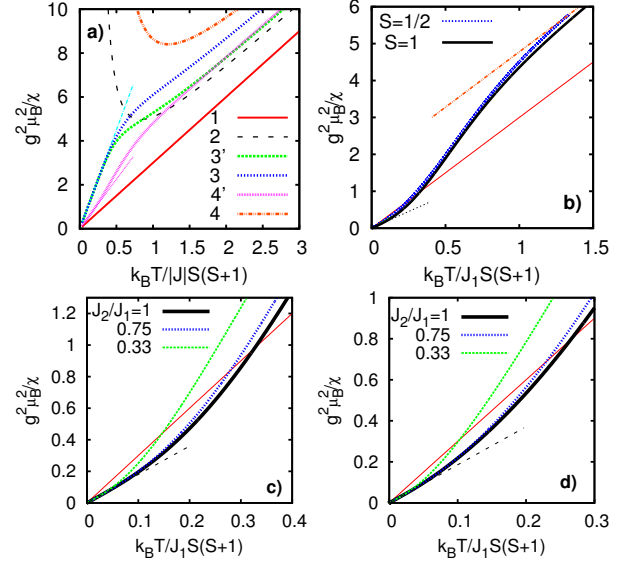


FIG. 2. (Color online) Inverse susceptibility (per spin)  $\chi^{-1}$  for the complexes shown in Fig. 1. Straight solid red line shows the Curie law  $\chi_C^{-1}$ ; straight dashed lines show the low-temperature asymptotics: "super" -paramagnetic Curie laws  $(g\mu_B)^2/\chi_g = 3Nk_B T/[S_g(S_g+1)]$  for Lieb-Mattis systems. **a)** : clusters shown in Fig. 1a with  $S = 1/2$ ; **b)** : the complex shown in Fig. 1b with two different values of spin  $S$ ; the straight dash-dotted red line is the high- $T$  Curie-Weiss asymptote. **c)** the same complex with  $S = 1$  and various values of  $J_2/J_1$ ; **d)** the complex shown in Fig. 1c with  $S = 1/2$  and various values of  $J_2/J_1$ .

in the ground state, in the absence of a long-range magnetic order<sup>19</sup>. We have performed full exact diagonalization studies (ED) of thermodynamic properties of clusters shown in Fig. 1b,c using J. Schulenburg's *spinpack* program<sup>20,21</sup>. The susceptibility  $\chi(T)$  is calculated as the ratio of the induced magnetization  $M$  to the "vanishing" magnetic field  $H = 10^{-5} J_1/g\mu_B$ . One observes in Figure 2b,c,d that the response of the systems shown in Fig. 1b,c *exceeds* the response of non-interacting spins at low temperature. Thus, an antiferromagnetic interaction may result in an *enhancement* of magnetic response if the geometry of spin arrangement favors the formation of a ferrimagnetic ground state. Then for temperatures  $T \ll T_s$  the susceptibility per spin shows superparamagnetic response  $\chi_g = [(g\mu_B)^2 S_g(S_g+1)]/[3k_B T(N_A + N_B)]$ . Evidently, the enhancement of the low-temperature response takes place, if

$$K \equiv \frac{\chi_g}{\chi_C} = \frac{|N_A - N_B|(|N_A - N_B|S + 1)}{(N_A + N_B)(S + 1)} > 1. \quad (3)$$

Not every system satisfying the requirements of the Lieb-Mattis theorem and having a ferrimagnetic ground state has an enhanced susceptibility. Thus, the clusters **3'** ( $N_A = 1, N_B = 2$ ) and **4'** ( $N_A = 1, N_B = 3$ ) both have  $K < 1$ , i.e. their response is weaker than that of the same number of non-interacting spins.

The "S"-shape form of the  $T$ -dependence of the inverse

susceptibility (Figure 2b) was previously reported for small fragments of ferrimagnetic superstructure in double perovskites<sup>10,22</sup>. It interpolates between the Curie-Weiss law  $\chi_{CW}$  at  $T \gg T_s$ , and the "super"-spin Curie law  $\chi_g = K\chi_C$  at  $T \ll T_s$ .

If impurity spins arrange themselves in a periodic superstructure having two (or more) non-equivalent spin positions, a ferrimagnetic ground state is possible for this superstructure. Let us denote the number of spins in the superstructure unit cell  $n_A + n_B$ , where A and B refer to the non-equivalent positions. If the spins of the sublattice A interact (antiferromagnetically) only with the spins of the sublattice B (absence of frustration), and  $n_A \neq n_B$ , the ground-state spin of the unit cell is  $S_c = S|n_A - n_B|$ <sup>8</sup>. For a fragment of such a ferrimagnetic superstructure

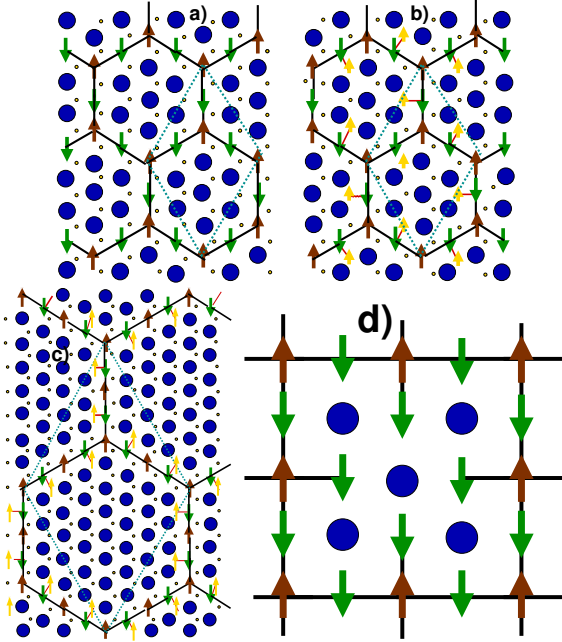


FIG. 3. (Color online) Examples of ferrimagnetic superstructures **a)**, **b)** : flat and three-dimensional two-leg honeycombs,  $L = 1$ ; **c)** : four-leg honeycomb,  $L = 2$ ; **d)** : a unit cell of a square network, it may be also regarded as a face of cubic unit cell. The notations is the same as in Fig. 1. The cyan rhombi show the unit cells.

containing  $N_c$  cells, the ground-state spin is  $S_g = N_c S_c = N_c |n_A - n_B| S$ , and the enhancement ratio equals  $K = |n_A - n_B| (N_c |n_A - n_B| S + 1) / [(n_A + n_B)(S + 1)]$ . It is clear that for a sufficiently large number of cells  $N_c$  the ratio  $K$  will be not only greater than 1, but can reach very large values. Fig. 3a shows a honeycomb superstructure that may be formed by TMIs in the  $ab$  plane of the wurtzite structure. The hexagon edge length is  $a_h = 2a$ ,  $a$  being the lattice parameter of the wurtzite. It is easy to imagine superstructures with  $a_h = 2La$ ,  $L = 1, 2, \dots$ , all of them being ferrimagnetic.

Flat superstructures like those shown in Fig. 3a can be linked together by some bridging spins to form a three-dimensional ferrimagnetic superstructure, which will un-

dergo a ferrimagnetic phase transition, provided that the number of cells is macroscopically large. Figure 3b,c

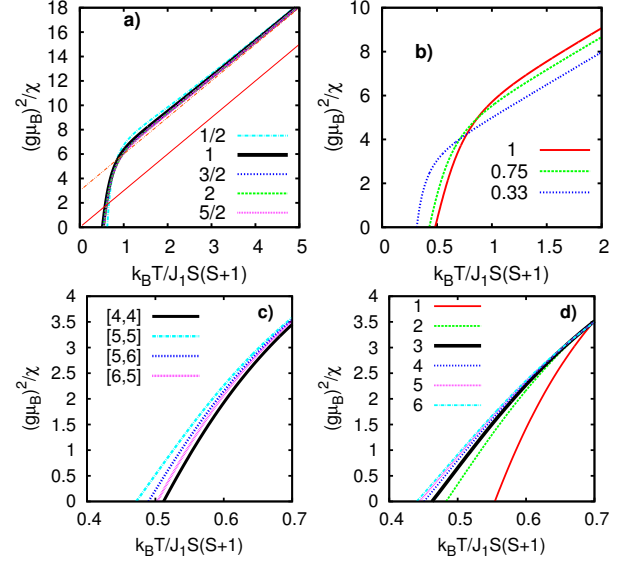


FIG. 4. (Color online) Temperature dependence of inverse susceptibility given by [5,5] Padé approximants for tenth-order high-temperature expansion (HTE) for ferrimagnetic superstructures: **a)** two-leg honeycomb ( $L = 1$ ), various spin values are shown, solid (dash-dotted) straight red line shows Curie (Curie-Weiss) law; **b)** four-leg honeycomb ( $L = 2$ ),  $S = 5/2$  various  $J_2/J_1$  values are shown; **c)** four-leg system,  $L = 2$ ,  $S = 2$  various Padé approximants for eighth-order ([4,4]<sup>23,24</sup>, tenth-order ([4,6], [5,5], [6,4])<sup>25</sup>, and eleven-order ([5,6], [6,5])<sup>26</sup> HTE; **d)** the vicinity of  $T_C$  for various honeycomb superstructures with size parameter  $L = 1, 2 \dots 6$ ,  $S = 5/2$ ,  $J_2 = J_1$ .

shows examples of the structures. It is clear that this motif may be repeated in an infinite number of variations. Like the host wurtzite lattice, the unit cell of the superstructure contains metal ions in two planes. The magnetic ions in one plane (green "down" and brown "up" arrows) form a honeycomb lattice with the hexagon edge  $2La$ . In the second plane, the magnetic ions (gold "up" arrows) occupy the positions nearest to the green "down" arrows. The interaction between the ions in the first plane is  $J_1$ , whereas the interaction between the ions in two adjacent planes is  $J_2$ . We note that the complexes shown in Fig. 1b,c are building blocks of the honeycombs. It will be demonstrated below that many other Lieb-Mattis networks can be built of such blocks. The number of magnetic ions in the unit cell is  $n_A + n_B = 9L - 1$ , the ground state spin of the cell being  $S_c = S|n_A - n_B| = S(3L - 1)$ . Now the total number of ions in the cell is  $n_c = 24L^2$ . Thus, the concentration of magnetic ions equals  $x = (9L - 1)/(24L^2)$ , and can be made very small for sufficiently large  $L$ . At the same time, the average ground-state spin per magnetic ion,  $\langle S_{\mathbf{R}} \rangle = S_c/(n_A + n_B) = S(3L - 1)/(9L - 1)$ , tends to a finite value,  $S/3$ , as  $L \rightarrow \infty$ .

The inverse magnetic susceptibility  $\chi^{-1}$  of such super-

structures is presented in Fig. 4 as a function of normalized temperature  $T/T_s$ . It was calculated using a program<sup>25</sup> based on the tenth-order high-temperature expansion (HTE)<sup>27</sup>. The program computes the exact coefficients of the HTE as well as its Padé approximants (ratios of two polynomials),  $\chi(T) \approx [m, n] = P_m(T)/P_n(T)$ . The Padé approximants allow to extend the region of validity of the HTE down to  $T \sim 0.5T_s$ <sup>25</sup> (Fig. 4c). This extension sometimes fails if an approximant has a pole in the temperature region of interest. Our experience shows that the [5,5] approximant works well in almost all cases. Sometimes difficulties arise for  $S = 1/2$ , and for small  $J_2/J_1$  ratios, i.e., for the extreme quantum case. Nevertheless, due to the weak dependence of the shape of the curve  $\chi^{-1}(T/T_s)$  on the spin value  $S$  (Fig. 4a), it can still be analyzed. At  $T \gtrsim 3T_s$ , the inverse susceptibility follows the Curie-Weiss asymptotic law with  $\theta = -[S(S+1)/3k_B]12L(J_1 + J_2)/(9L - 1)$ . For  $T \lesssim T_s$  it sharply deviates from the asymptotic behavior and changes sign at  $T = T_C$ . This is the temperature of ferrimagnetic ordering — the Curie temperature.

The precision of the determination of critical temperatures from the zero of  $\chi^{-1}$  (Fig. 4c) was estimated to be about 10%<sup>25</sup>. Figure 4b shows that  $T_C$  decreases as the ratio of out-of-plane to in-plane couplings,  $J_2/J_1$ , is reduced. At  $J_2 = 0$  the system becomes a stack of non-interacting two-dimensional planes, and  $T_C$  should vanish. This limit lies outside the range of applicability of the HTE, and we postpone its study to future works. Here we mention only that magnetic anisotropy, which is neglected in our study, should act in the opposite direction, i.e., it should enhance the  $T_C$  as it depresses spin fluctuations.

Figure 4d shows that the ordering temperature decreases very slowly as  $L$  is increased. Note that the superstructure parameter values  $L = 1, 2, 3, 4, 5, 6$  correspond to the following concentrations of magnetic ions:  $x = 0.33, 0.18, 0.12, 0.09, 0.07, 0.06$ . To get a closer relation to experiments, we may consider, e.g., ZnO:Mn,Co, where the in-plane superexchange values are  $J_1/k_B \sim 50$  K<sup>11,13,14</sup> and  $T_s = J_1 S(S+1)/k_B \sim 438(188)$  K for  $S = 5/2(3/2)$ . For other Co-doped semiconductors  $66$  K  $\lesssim J_1/k_B \lesssim 100$  K<sup>12,17,28</sup> (and references therein), i.e.,  $T_s$  lies within the interval  $248$  K  $\lesssim T_s \lesssim 375$  K. The Mn-doped semiconductors have  $12$  K  $\lesssim J_1/k_B \lesssim 32$  K<sup>12,29</sup>, and  $105$  K  $\lesssim T_s \lesssim 280$  K.

Thus, a very diluted system may have an appreciable ordering temperature ( $T_C \gtrsim 100$  K) provided that the magnetic ions are arranged in a Lieb-Mattis ferrimagnetic superstructure.

In many aspects, the behavior of a ferrimagnet in its ordered state is similar to that of a ferromagnet with the same value of spontaneous magnetization  $M_s$ . But in a high magnetic field the ferrimagnet exhibits a transition accompanied by reorientation of its sublattices<sup>30–32</sup>. At  $T = 0$  the magnetization per spin has a constant value,  $\mu_{M,s} = g\mu_B S|n_A - n_B|/(n_A + n_B)$ , up to a certain critical field,  $H_{c,1}$ ; then it grows up linearly to the

saturation value,  $\mu_{M,max} = g\mu_B S$ , which is reached at a second critical field,  $H_{c,2}$ . For a two-sublattice ferrimagnet having the structure shown in Fig. 3a ( $L = 1$ ) and  $J_1 = J_2 = J$  we find  $g\mu_B H_{c,1} = JS$ , and  $H_{c,2} = 5H_{c,1}$ . For  $J/k_B \sim 20$  K this gives  $H_{c,1} \sim 37$  T,  $H_{c,2} \sim 185$  T.

The complexes shown in Fig. 1b,c may be arranged in many kinds of networks, to form Lieb-Mattis ferrimagnetic superstructures in various host semiconductors. Figure 3d shows an example of a 2D square superstructure unit cell with  $L = 2$ , which is possible in a cubic host. It has  $n_A = 1 + 2(L - 1)$  and  $n_B = 4(L - 1) + 2L$ . One can also imagine a 3D cubic network; then Fig. 3d corresponds to a face of the cubic unit cell having  $n_A = 1 + 3(L - 1)$ ,  $n_B = 3L + 12(L - 1)$ , and the concentration of magnetic ions  $x = (n_A + n_B)/n_c = (9L - 7)/(4L^3)$ . Formation of such superstructures is possible in perovskite solid solutions, like  $\text{KMn}_x\text{Mg}_{1-x}\text{F}_3$ <sup>33,34</sup>, or in solutions of multiferroics  $\text{PbFe}_{1/2}\text{Nb}_{1/2}\text{O}_3$  or  $\text{PbFe}_{1/2}\text{Ta}_{1/2}\text{O}_3$  with ferroelectric perovskites<sup>35–39</sup>.

We conclude that Lieb-Mattis ferrimagnetism is a possible route to obtaining long-range magnetic order in semiconductors containing transition metal ions as substitutional impurities, which requires no additional charge carriers. A precursor of the ordering transition is the enhanced magnetic response of finite cluster showing isotropic superparamagnetism. Our results for the inverse susceptibility show a characteristic "S"-like form of the curves, which could be used to identify the present mechanism. Adding the magnetic anisotropy to our theory, we expect also other ingredients of superparamagnetism, namely a finite blocking temperature and hysteresis.

These superparamagnetic clusters serve as building blocks to create infinite sublattices of the wurtzite structure that obey the Lieb-Mattis rules. As we have already noted, there is an enormous wealth of such Lieb-Mattis sublattices, our proposals (Fig. 3) may only serve as examples. We expect a finite transition temperature for all these lattices and we have shown it explicitly for the subclass that we considered. Of course, a question arises, whether frustration in a realistic diluted semiconductor can influence the above discussed scenario. First we argue that there are several numerical studies showing that the Lieb-Mattis theorem, although not rigorously valid, applies to many frustrated spin systems, see, e.g., Ref. 40. Furthermore, we know that there are various frustrated 2D lattices with antiferromagnetic nearest-neighbor exchange, such as the triangular or the Shastry-Sutherland lattices, which show ground-state magnetic LRO<sup>41,42</sup>. Last but not least, the stability of the ferrimagnetic ground state against frustration has been demonstrated for several specific ferrimagnetic models, see, e.g., Refs. 43–45. Consequently, there is ample evidence that the above sketched mechanism should be robust against frustration. The final proof that the here proposed mechanism can, indeed, be realized in a real material demands further studies, in close collaboration between experiment and theory.

In this communication, we have considered only semiconductors doped by one kind of magnetic ions, where ferrimagnetism can appear due to the topology of interacting bonds. Another option is the co-doping with two kinds of ions having different spin values. In both cases a ferrimagnetic semiconductor may be a good alternative

to a ferromagnetic one.

## ACKNOWLEDGMENTS

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